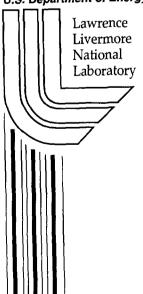
## Natural-Gas-Assisted Steam Electrolysis for Distributed Hydrogen Production

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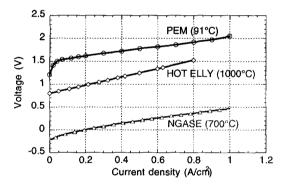
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Due to greenhouse gas emission concerns, hydrogen has become increasingly important as a potential clean fuel for transportation. Since the infrastructure for hydrogen production and delivery is almost non-existing, there is a clear need for the development of an efficient distributed hydrogen production technology. Currently, most of the total hydrogen demand is met by hydrogen production from fossil fuels, i.e., by steam reforming of natural gas and by coal gasification. Hydrogen can be produced from water electrolysis using simpler technology with reduced local environmental pollution. Besides simplicity, electrolysis has also modular characteristic, which makes on-site production of hydrogen extremely attractive. However, water electrolysis has not had significant commercial impact because it has not been possible to make it cost effective. For example, the electrical consumption necessary for hydrogen generation in the German HOT ELLY process (using steam electrolysis) represents 80% of the total production cost (1). The resulting cost per unit volume of hydrogen produced was found to be about twice that of steam-reformed hydrogen, prohibiting commercialization and widespread utilization of steam electrolyzers.

At Lawrence Livermore National Laboratory (LLNL), we have developed a novel approach to reduce the electricity consumption in solid oxide steam electrolyzers (2). The principal electrical efficiency gain is realized by minimizing the open-circuit voltage against which the steam electrolyzer is forced to operate. To accomplish this, natural gas is used to depolarize the anode in the socalled Natural-Gas-Assisted Steam Electrolyzer (NGASE). There are two possible operational modes for the NGASE. In a total oxidation mode, the oxygen coming from electrolysis in the cathode side is used to completely oxidize methane to CO<sub>2</sub> and H<sub>2</sub>O, which is analogous to the reactions occurring in a solid oxide fuel cell. The total energy required for water splitting is equivalent to that required in a conventional electrolyzer where oxygen is exhausted into the environment. However, replacement of part of the electricity requirement by an equivalent (energy) amount of natural gas at one-fourth the cost will significantly reduce the hydrogen production cost. The NGASE efficiency with respect to primary energy will be significantly higher than that of conventional electrolyzers, due to the fact that electricity required for conventional electrolysis must be generated by burning coal or natural gas at an average efficiency below 40%. In the partial oxidation mode, methane will be converted to CO and H2. CO can then be subsequently shifted to vield more hydrogen. In this mode, for an equivalent amount of current passing through the electrolyzer, the hydrogen production will be four times greater than in conventional electrolyzer systems. Current research is focused on the total oxidation mode because of its simplicity.

A single cell electrolyzer was prepared and tested. The anode, made of Ni/yttria-stabilized-zirconia (YSZ) cernet, serves as support for the electrolysis cell. A YSZ thin film electrolyte was deposited using the Colloidal Spray Deposition technique (3). The thin film cathode, made of Ni/YSZ or Ni/yttria-doped-ceria cermet, was also deposited using colloidal deposition. Cell testings were done in wet hydrogen in the cathode side and wet methane in the anode side at various temperatures and various steam concentration.

Figure 1 shows the current-voltage characteristics of the NGASE cell as well as those for the conventional polymer electrolyte membrane (PEM) electrolyzer and for the HOT ELLY steam electrolyzer. Due to the higher operating temperature, the HOT ELLY voltage is lower than that of the PEM electrolyzer, resulting in lower electrical consumption. The NGASE cell has even lower voltage. The open-circuit voltage actually becomes negative and the reaction is driven spontaneously in a similar way as a fuel cell. The gain in voltage reduction is as high as 1 V when methane is used to depolarize the anode. The use of thin film and of high performance electrode has made possible to lower the electrolyzer operating temperature to 700°C. This operating



temperature is favorable to the total oxidation of the methane.

A tubular electrolyzer stack is currently under development.

Figure 1. I-V characteristics of PEM, HOT ELLY and NGASE electrolyzers.

## References

- 1. K.H. Quandt and R. Streicher, *Int. J. Hydrogen Energy* 11, 309 (1986).
- $2.\ A.Q.$  Pham, H. Wallman and R.S. Glass, US patent 6051125.
- 3. A.Q. Pham, T.H. Lee, B. Chung and R.S. Glass, "Colloidal Spray Deposition Technique for the Processing of Thin Film Solid Oxide Fuel Cells", in Proceedings of the 6<sup>th</sup> International Symposium on Solid Oxide Fuel Cells, edited by S. C. Singhal and M. Dokiya, The

Electrochemical Soc., Pennington, NJ. USA (1999) p.172.

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